DOCKET NO. 146800

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Serial Number.	10/655,988
Application to Reissue U.S. Pat.	5,405,911
Filed:	Sept. 4, 2003
Inventor:	Handlin, Jr., et al.

For: Butadiene Polymers Having Terminal Functional Groups

Notice of Reissue Application Published in O.G.: December 23, 2003

DECLARATION IN SUPPORT OF PROTEST

I, Roderic P. Quirk, hereby depose and say that:

1. I am currently a Distinguished Professor of Polymer Science and the Kumho Professor of Polymer Science in the Department of Polymer Science at the University of Akron.

2. I have been a Professor of Polymer Science at the University of Akron for 20 years.

3. I have been teaching and doing research in polymer science for 30 years.

4. My main research area in the synthesis and characterization of polymers, especially chain-end functionalized polymers, using anionic polymerization.

5. I have published more than 200 scientific papers and I am the inventor or co-inventor on 28 patents.

6. I have also co-authored a book on anionic polymerization and edited five other books.

7. I have carefully read the patents of D. L. Handlin, D. E. Goodwin, C. L. Willis, D. J. St. Clair, J. D. Wildy, M. J. Modie and C. A. Stevens (U.S. 5,405,911 and 5,393,843) and have been requested to comment on whether or not the claims made by the inventors are obvious to one who is skilled in the art.

8. Handlin et al. claim that the melt viscosities of the hydrogenated, telechelic, dihydroxy polybutadienes are unexpectedly small compared to other analogous, commercial polymers. However, the analogous, commercial polymers all have a different microstructure (content of comonomer units) along the polymer chain.

9. The Handlin et al. polymers have microstructures that correspond to 50-52 % ethylethylene units, while the allegedly analogous commercial polymers have ca. 85 % ethylethylene units.

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10. For both polymers, the remainder of the comonomer units correspond to ethylene units.
Since well before the filing date of the Handlin et al. patent application, it has been well
known to polymer scientists skilled in this art that the viscosity of a polymer depends on the
temperature at which the viscosity is measured relative to the polymer glass transition
temperature. This was established in 1955 by the seminal paper by Williams, Landel and
Ferry [M. L. Williams, R. F. Landel, J. D. Ferry, Journal of the American Chemical Society,
vol. 77, p. 3701(1955)]. The relationship between these quantities is embodied in the well-
known WLF equation. The viscosity is higher when the measurement temperature is close to
the glass transition temperature of a polymer and it decreases as the measurement
temperature increases compared to the glass transition temperature. For example, the WLF
relationship is described in the polymer textbook of Sperling ("Introduction to Physical
Poymer Science", L. H. Sperling, Wiley-Interscience, New York, 1986, p. 261) where T_g is
the glass transition temperature, η is the viscosity at temperature T, η_g is the viscosity at the
T_g.

$$\text{Log } (\eta/\eta_g) = -17.44(T-T_g)/51.6 + T-T_g$$

For a reference temperature set to an arbitrary temperature, T_s,
(e.g. 50 °C above T_g), the Williams, Landel, Ferry (WLF) constants provide the following
equation where T_s is the reference temperature, η is the viscosity at temperature T, η_s is the
viscosity at the reference temperature.

$$\text{Log } (\eta/\eta_s) = -8.86(T-T_s)/101.6 + T-T_s$$

These equations embody fundamental relationships that are known to one skilled in the art of
polymer science. These equations show that the viscosity decreases as the temperature is
increased above the glass transition temperature. Furthermore, these equations are general,
i.e., they do not depend on the structure of the polymer other than these relationships are
general for amorphous polymers.

12. The consequences of these relationships are clear also. If one compares viscosities for
different polymers of a family of polymers at the same temperature, the polymer that has a
higher glass transition temperature (i.e., a T_g closer to the measurement temperature) will

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exhibit a higher viscosity. However, if the viscosities of these same two polymers in a family of polymers are both measured at the same temperature interval above their respective glass transition temperatures (e.g. $T-T_g$), they will exhibit the same viscosities. Therefore, if two polymers in a family of polymers have different glass transition temperatures, they would be expected to exhibit different viscosities at some arbitrary measurement temperature (e.g., at 24 •C as described in U.S. 3,629,172).

13. These fundamental principles have been well known for many years (e.g. since 1955) to polymer scientists skilled in this art and working with viscosities of polymers. These concepts have been thoroughly and clearly presented in the classic textbook by John D. Ferry, "Viscoelastic Properties of Polymers", 3rd ed., Wiley, 1980. It is noteworthy that the first edition of this text appeared in 1961.

14. With respect to the glass transition temperatures of the polymers in question, it is useful to consider the two extremes. Polyethylene would correspond to a hydrogenated polybutadiene with no ethylethylene branches. Although this polymer would be expected to be semi-crystalline, the estimated T_g for this hypothetical polymer is – 80 ° C (see R. F. Boyer, "Transitions and Relaxations" in Encyclopedia of Polymer Science and Technology, Supplement No. 2, Wiley, New York, 1977, p. 812). For the polymer at the other extreme of the structure spectrum, polyethylethylene [i.e. also referred to as poly(1-butene)] has a T_g of ca. –33 °C (see Handbook of Polymer Handbook, 4th Edition, J. Brandrup, E. H. Immergut, E. A. Grulke, Eds, Wiley-Interscience, New York, 1999, p. V206). In general the T_g of copolymers varies in a systematic way depending on the composition and T_g of the comonomers as expressed by the Fox equation (see Sperling, loc. cit., p. 277) shown below

$$1/T_g = (M_1/T_{g1}) + (M_2/T_{g2})$$

where T_g is the glass transition temperature of the copolymer with the composition of mass M_1 of comonomer 1 and mass M_2 of comonomer 2 and T_{g1} is the glass transition temperature of the pure homopolymer of monomer 1 and T_{g2} is the glass transition temperature of pure monomer 2. This equation predicts a linear relationship between composition of the copolymer and the glass transition temperature of the copolymer. Once again, this relationship has been common knowledge to those skilled in the art of polymer science for many years.

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15. From the Fox equation and the glass transition temperatures of the two homopolymers [polyethylene $T_g \approx$ -80 °C and polyethylethylene with $T_g \approx$ -33 °C], it is straightforward to predict that the glass transition temperatures of the family of ethylene/ethylethylene copolymers (i.e., the products of hydrogenation of polybutadienes with different amounts of 1,4 and 1,2-microstructures) would increase with increasing amounts of ethylethylene units. Thus, the analogous commercial polymers with higher contents of ethylethylene units would have higher T_g values. Correspondingly, these commercial polymers would also be expected to have higher viscosities relative to the polymers in question in the Handlin et al. patent because the Handlin polymers have lower contents of ethylethylene units and therefore would be expected to have lower T_g values and lower viscosities. As a consequence of these fundamental principles that should be well known to anyone skilled in the art in polymer science, the observations of lower viscosities for the polymers in the patent relative to the allegedly analogous commercial polymers is not unexpected; in fact, it is expected based on basic principles of polymer science, i.e., the WLF equation.

16. The effect of chain microstructure on the viscoelastic properties of the family of hydrogenated polybutadienes and the applicability of the WLF relationship to correlate these data have been described by J. M. Carella, W. W. Graessley, F. J. Fetters, Macromolecules, vol. 17, p. 2775(1984).

April 20, 2004
Date

Roderic P. Quirk

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FILE NAME: PROTEST TO REISSUE OF US. PAT 5,405,911 - KRATON MATTER

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Following is: Supplement to Protest on 3 pages plus Declaration of Dr. Roderic Quirk in Support of Protest on -4 pages -- including this cover sheet 9 pages

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